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GUSTAV MIE AND THE EVOLVING DISCIPLINE OF ELECTROMAGNETIC SCATTERING BY PARTICLES

BY MICHAEL I. MISHCHENKO AND LARRY D. TRAVIS

Despite being a century old, the Mie theory keeps revealing secrets of electromagnetic scattering by particles and helps develop new theoretical methods and advanced remote sensing and in situ particle characterization techniques.

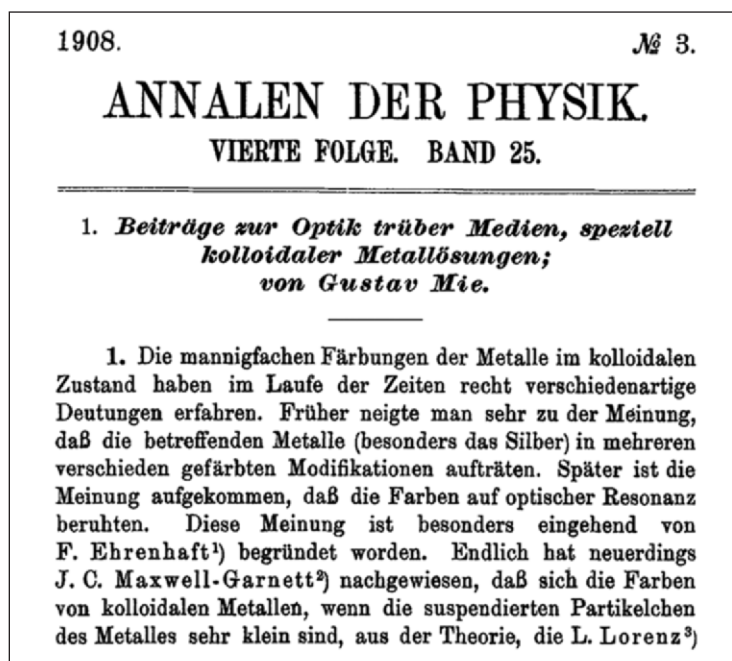


FIG. 1. The first page of Mie's 1908 paper.

Gustav Mie's paper under the title "Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen" (Contributions to the optics of turbid media, particularly colloidal metal suspensions) appeared in 1908 (Fig. 1) as part of the third issue of the 25th volume of the renowned German physics journal *Annalen der Physik* (Mie 1908). This paper took aim at a theoretical explanation of the beautiful coloration of metals in a colloidal state and was triggered by an experimental dissertation of Walter Steubing, a student of Mie's at the Physical

Institute of Greifswald. At that time Mie considered his treatise to be a rather trivial application of Maxwell's electromagnetics. He simply did not anticipate the eventual phenomenal success of this paper and universal acceptance of his exercise in mathematical physics as "the Mie theory."

For 100 yr the Mie theory has been an indispensable tool in the development of electromagnetic scattering theory as well as computational and measurement techniques and methodologies. Since the advent of computers in the 1940s, the number and

diversity of practical applications of the Mie theory in such disciplines as climate modeling, optical particle characterization, astrophysics, nanoscience, and biomedical optics have been astounding. Undoubtedly, this can be attributed to the ubiquity and universal importance of electromagnetic scattering by particles coupled with the unparalleled simplicity, accuracy, and efficiency of the Mie theory and its virtually unlimited richness. In particular, one cannot even imagine the modern-day functioning of such disciplines as atmospheric radiation and remote sensing without the Mie theory: the overwhelming majority of calculations of electromagnetic scattering and radiative transfer in the atmosphere, ocean, and particulate surfaces is now directly based on Mie computer codes.

By now Mie's 1908 paper has been cited in almost 4,000 journal articles since 1955 (according to the inherently incomplete Science Citation Index Expanded database), and the citation rate appears to increase rather than decrease with time. This magnitude of success is highly unusual for a seemingly dry, abstract, and specialized article on physics and definitely places Mie's paper in the category of one of the more influential scientific publications of the twentieth century. It thus appears highly appropriate to celebrate the centenary of the seminal Mie paper by analyzing its virtues and importance and by placing it in a broader evolving context of electromagnetic scattering by particles.

THE MIE THEORY AND THE CONCEPT OF ELECTROMAGNETIC SCATTERING. The fundamental nature of Maxwell's electromagnetics is now universally recognized. In a recent poll of scientists, the Maxwell equations have been voted to be the "greatest equations ever . . ." (Crease 2004). However, the situation in the late nineteenth and early twentieth centuries was somewhat different. While one of the great "continental" physicists Ludwig Boltzmann had

immediately recognized the universal importance of the Maxwell equations and was even quoted, "Was it god who wrote these lines . . ." (Sommerfeld 1952), the stance of many other continental and even British physicists had not been so unequivocal.

One of the decisive virtues of the Mie paper happened to be its explicit reliance on Maxwell's electromagnetics. As such, this paper would eventually be recognized as one of the great triumphs of the Maxwell theory. It is now widely acknowledged that the brilliant Danish physicist Ludvig Valentin Lorenz (1829–91) developed a theory of light scattering by spherical particles, which is mathematically very similar to the Mie theory (Logan 1965; Kragh 1991). Yet he based his memoir on his own theory of light, which was at variance with the Maxwell theory in that it was based on the concept of a light vector rather than on the concept of electromagnetic field. This made the physical interpretations of the scattering theories of Mie and Lorenz radically different and has ultimately led to the neglect of Lorenz's contribution. Another important ingredient of the Mie theory differentiating from Lorenz's work was the explicit assumption of a potentially absorbing host medium.

The Mie theory explicitly deals with electromagnetic scattering by homogeneous spherical particles. In modern physical terms, this theory belongs in the realm of so-called frequency-domain macroscopic electromagnetics. This means, in particular, that all fields and sources of fields are assumed to vary in time harmonically [i.e., are proportional to the common factor $\exp(i\omega t)$, where $i = (-1)^{1/2}$, ω is the angular frequency, and t is time]. The fundamental concept of electromagnetic scattering used by Mie can be illustrated as follows: a plane electromagnetic wave propagates in an infinite nonabsorbing medium without a change in its intensity or polarization state (Fig. 2a). However, the presence of a particle modifies the electromagnetic field that would otherwise exist in the unbounded homogeneous space. It is this modification that is called *electromagnetic scattering*. The difference between the total field in the presence of the particle (Fig. 2b) and the original field that would exist in the absence of the particle can be thought of as the field scattered by the particle (Fig. 2c). In other words, the total field in the presence of the particle is represented as the vector sum of the respective incident (original) and scattered fields: $\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \mathbf{E}^{\text{sca}}(\mathbf{r})$, where \mathbf{r} is the position vector and the common factor $\exp(i\omega t)$ is omitted. It should be recognized that the division of the total field into the incident and scattered parts is a purely mathematical procedure. This means that classical frequency-domain electromagnetic scattering is

AFFILIATIONS: MISHCHENKO AND TRAVIS—NASA Goddard Institute for Space Studies, New York, New York

CORRESPONDING AUTHOR: Dr. Michael I. Mishchenko, NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025

E-mail: mmishchenko@giss.nasa.gov

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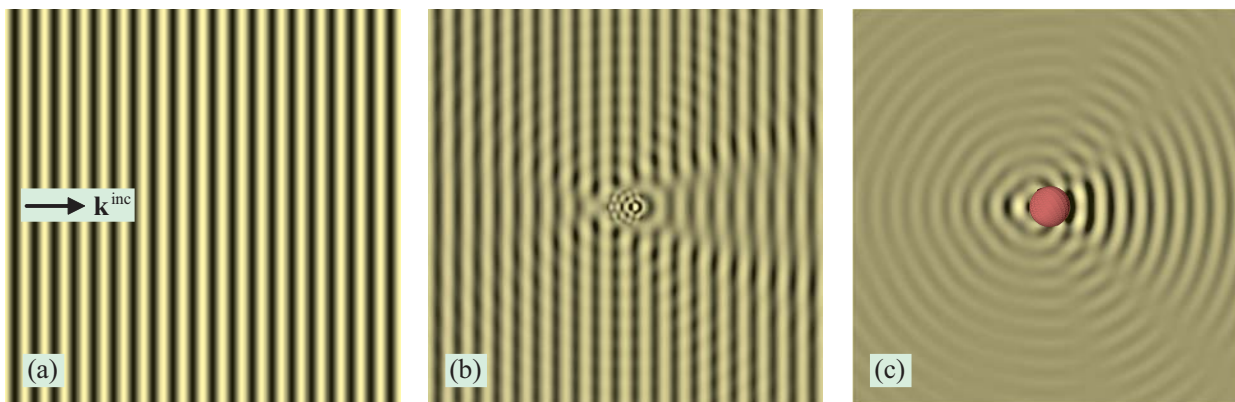


FIG. 2. (a) The real part of the vertical (i.e., perpendicular to the paper) component of the electric field vector of a plane electromagnetic wave propagating in the direction of the wave vector \mathbf{k}^{inc} . The infinite host medium is homogeneous, isotropic, and nonabsorbing. The wave is fully polarized in the vertical direction so that the horizontal component of the electric field vector is equal to zero. (b) The real part of the vertical component of the total electric field in the presence of a small homogeneous spherical particle located in the center of the diagram as to be shown in (c). The relative refractive index of the particle is 2.8, while its radius is equal to the wavelength. (c) The real part of the vertical component of the difference between the fields visualized in (b) and (a). The color scale is individually adjusted to maximally reveal the specific details in each diagram.

not a physical process per se but rather an abbreviated way to state that the total field computed in the presence of a particle is different from that computed in the absence of the particle. In other words, frequency-domain electromagnetic scattering is a physical phenomenon, but not a physical process.

This concept of electromagnetic scattering by a particle remains as valid now as it was 100 yr ago. Yet it is truly remarkable how many confusing and even plainly wrong definitions of scattering have appeared in the literature since the publication of Mie's paper. Despite the purely classical character of scattering of *waves* in the framework of macroscopic frequency-domain Maxwell electromagnetics, one may frequently encounter the assertion that upon *collision* with an atmospheric particle, the incident *photon* can be either absorbed or scattered. Scattering is then defined as a random choice of new direction of propagation for the photon *according to the Mie theory*.

The neo-Newtonian visualization of scattering as a “collision” of a light corpuscle with a cloud droplet followed by the corpuscle changing the direction of flight appears to be intuitively appealing and is rather common. However, this artificial association of photons and the Mie theory invariably falls apart upon a closer look at what is actually meant by a “photon.” Indeed, although the physical concept of a photon is a valid one, it appears and must be used only in the rigorous context of quantum electrodynamics (Meystre and Sargent 1999). Otherwise the phenomenological association of the real physical photons

with “particles of light” creates profound confusion as discussed by Kidd et al. (1989) and Lamb (1995). Obviously, it is technically impossible to explicitly quantize the electromagnetic field in the presence of a material body (such as the cloud droplet) consisting of an enormous number of elementary particles. However, the electromagnetic scattering problem can be solved self-consistently by first deriving macroscopic electromagnetics from quantum theory and statistical physics (Akhiezer and Peletminskii 1981) and then solving the macroscopic Maxwell equations: hence the great heuristic value and practical usefulness of the Mie theory.

Mie intentionally constructed his solution of the Maxwell equations in such a way that the scattered field transforms into an outgoing spherical wave at a sufficiently large distance from the particle, in the so-called far-field zone (see Fig. 2c). This far-field assumption was a brilliant insight made largely on “physical grounds” and was intended to model situations involving widely separated particles such as cloud droplets. However, the general behavior of the scattered field at infinity later turned out to be at the very heart of the fundamental problem of uniqueness of solution of the Maxwell equations. In fact, in constructing his solution Mie anticipated what is now called the Sommerfeld–Silver–Müller radiation condition at infinity applicable to electromagnetic scattering by an arbitrary finite object imbedded in a nonabsorbing, unbounded, homogeneous medium. This condition requires the longitudinal components of the electric and magnetic fields to decrease at infin-

ity faster than the transverse components. Then it can be proven mathematically that the Maxwell equations have a unique (and hence physically relevant) solution. This fundamental aspect of electromagnetic scattering is discussed thoroughly in Müller (1969).

While being quantitatively rigorous, the Mie theory is sometimes viewed as a “black-box” numerical tool for not always yielding as clear a “physical picture” of electromagnetic scattering as some may like. For example, while the Mie solution does yield the famous Rayleigh scattering law in the limit of vanishingly small particle radius, it has not been shown analytically to explain the equally famous “extinction paradox” according to which the extinction cross section of a large sphere is equal to twice its geometrical cross section. Potential limitations of this kind have stimulated research aimed at providing deeper qualitative insights into well-known optical phenomena such as the rainbow and the glory discussed below (e.g., Nussenzweig 1992). Hopefully such studies will eventually yield a predictive capability that will allow one to foresee scattering phenomena and effects so far obscured by the numerical intricacy of the Mie solution.

GENERALIZATIONS OF THE MIE THEORY.

The Mie theory belongs to the class of separation-of-variables solutions in that it explicitly exploits the separability of the vector Helmholtz equation for the time-harmonic electric field in polar spherical coordinates. A seminal theoretical development was the reformulation of the Mie theory by Stratton (1941) in terms of special so-called vector spherical wave functions (VSWFs) possessing very convenient analytical properties. In the framework of Stratton’s formulation, the electromagnetic scattering problem is solved by expanding the incident, internal, and scattered fields in appropriate sets of VSWFs. The expansion coefficients of the incident plane wave are computed analytically, while the unknown expansion coefficients of the internal and scattered fields are then determined through the requirement of the standard boundary conditions on the sphere surface as well as the radiation condition at infinity. Because the VSWFs are orthogonal on the sphere surface, the resulting formulas have the utmost simplicity.

The separation-of-variables approach affords a rather straightforward extension to simple models of inhomogeneous particles encountered in natural and laboratory environments such as concentric core-mantle spheres, concentric multilayered spheres, and radially inhomogeneous spheres. Furthermore, one can solve the scattering problem for a homogeneous or

layered spheroid using spheroidal coordinates. Such particles represent perhaps the simplest model of non-spherical aerosol and cloud particles. Unfortunately, in this case the expansion of the scattered field is a double series, and the solution turns out to be rather complicated analytically and time consuming when implemented on a computer.

Stratton’s reformulation paved the way to several direct generalizations of the Mie theory, all of which were proposed in the late 1960s to early 1970s and contain the original Mie theory as a particular case. Conceptually the simplest of them is the point-matching method (PMM), in which the expansion coefficients of the internal and scattered fields are determined through the requirement of the boundary conditions on the surface of a nonspherical scatterer. In principle, this technique is applicable to an arbitrarily shaped and sized particle. However, increasingly poor convergence of the simple original form of the PMM imposes a rather severe practical limit on the maximal deviation of the particle shape from that of a perfect sphere. This problem may have been ameliorated, at least partially, with more advanced and complex versions of the PMM.

The so-called extended boundary condition method (EBCM) is also quite general in principle, although it has been mostly applied to axially symmetric particles. Nevertheless, it has a wide practical range of particle sizes and aspect ratios and has been one of the most frequently used numerically exact techniques based on a direct solution of the Maxwell equations.

The multisphere or superposition *T*-matrix method is explicitly based on the so-called translation-addition theorem for VSWFs and was developed to treat electromagnetic scattering by clusters of spheres. Like EBCM, it has been used in a wide range of practical applications, most notably in computations of scattering and absorption properties of fractal soot aggregates resulting from fuel combustion. Similar mathematical properties of vector spheroidal wave functions permitted the development of a technique analogous to the superposition *T*-matrix method but intended to treat electromagnetic scattering by a cluster of spheroids in spheroidal coordinates. Specific information about all the above techniques and further references can be found in the collective monograph by Mishchenko et al. (2000) as well as in the review by Kahnert (2003).

The Mie theory is explicitly based on the assumption that the incident field is a plane electromagnetic wave. However, some types of illumination (e.g., a focused and/or very narrow laser beam) may

substantially violate this assumption. Still, the Mie solution of the Maxwell equations can be applied provided that the incident field is mathematically expandable in plane waves. A thorough review of this generalization of the Mie theory was given by Gouesbet and Gréhan (2000).

APPLICATIONS OF THE MIE THEORY.

Besides Stratton's book of 1941, an important role in the dissemination and popularization of the Mie theory has been played by the monographs by Born and Wolf (1959), Kerker (1969), and Bohren and Huffman (1983). However, the monograph by van de Hulst (1957), which in itself is one of the classical scientific treatises of the twentieth century, has had the greatest impact by far.

Despite being often characterized as an "analytical" solution, for most practical uses the Mie theory must be implemented on a computer. The great efficiency of modern desktop workstations and PCs coupled with the development of efficient algorithms for the computation of special functions entering the Mie solution (e.g., Wiscombe 1980) has made possible physically based applications involving massive calculations of electromagnetic scattering by particles. As a consequence, Mie codes have become an everyday tool for a large and rapidly expanding body of meteorologists, physicists, astrophysicists, and biologists as well as optical, electrical, and nano-science engineers. Furthermore, the Mie theory is now firmly established as a fundamental aspect of graduate and even undergraduate courses on atmospheric radiation and remote sensing (Stephens 1994; Liou 2002; Petty 2006). The remarkable reach of the Mie theory is well illustrated by a recent implementation of a small Matlab Mie scattering program on a Java-enabled mobile phone (Wriedt 2008). This allows the use of mobile phones as an active and portable educational tool, helping students quickly and interactively obtain a good working knowledge of the dependence of light scattering on particle size and refractive index.

We have already mentioned that the ability of the Mie theory to reproduce, both qualitatively and quantitatively, many actual observations and measurements of light scattering has been one of the great triumphs of Maxwell's electromagnetics. Among the most illustrative demonstrations of this ability have been accurate computer simulations of such spectacular natural optical displays as the primary and secondary rainbows produced by raindrops (Fig. 3) and the glory caused by cloud droplets (Fig. 4). Numerous applications of the Mie theory in meteorological optics and further useful links can be found online (www.philiplaven.com).

Figure 5 provides a vivid demonstration of the enormous richness of the Mie solution. Figure 5a corresponds to the case of monodisperse spheres and was created using moderate increments in both scattering angle and particle size parameter. It reveals a complex distribution of the areas of positive and negative polarization first displayed in this manner by Hansen and Travis (1974). This distribution resembles the pattern of spots on the wings of the Madagascan sunset moth (Yoshioka and Kinoshita 2007) and hence is called the "butterfly structure."

A zoomed-in polarization diagram shown in Fig. 5b reveals that some of the elements of the butterfly structure are extremely fine. They are associated with so-called super-narrow morphology-dependent



FIG. 3. Full-color numerical Mie simulation for 400- μm -radius water drops superimposed on a photograph of the primary and secondary rainbows. The Mie simulator used to create the artificial rainbows is described in Laven (2003) (image courtesy of P. Laven).

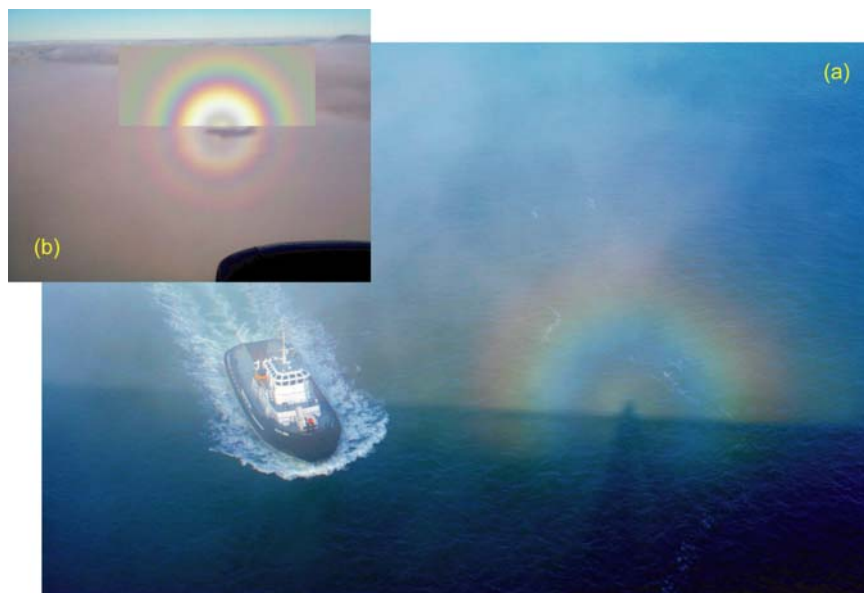


FIG. 4. (a) A beautiful glory photographed from San Francisco's Golden Gate Bridge and caused by swirling fog above cold water (photograph courtesy of L. Zinkova). (b) Full-color numerical Mie simulation of scattering of sunlight from 4.8- μm -radius water drops superimposed on the photograph of a glory taken from a commercial aircraft (image courtesy of P. Laven).

resonances (MDRs) discovered in the 1970s (Chýlek et al. 1978) and are caused by sharp minima in the denominators of analytical expressions for the Mie coefficients a_n and b_n as functions of size parameter, refractive index, or **radial structure**. Owing to the strong sensitivity of MDRs to particle physical pa-

rameters, laboratory and in situ measurements of resonance features in polarization and other scattering characteristics have been increasingly used for precise optical particle characterization (Davis and Schweiger 2002).

Averaging over a size distribution yields a much smoother polarization diagram, as shown in Fig. 5c. This diagram is more representative of natural and artificial polydisperse particles and was, in fact, used by Hansen and Hovenier (1974) in their analysis of ground-based polarimetric observations of Venus. As a result, they were able to determine with extreme precision the microphysical properties of cloud particles in the Venus atmosphere. The retrieved spectral behavior of the refractive index allowed an unequivocal identification of the particle chemical composition as a concentrated (76% by weight) aqueous solution of sulfuric acid. This spectacular achievement in planetary astrophysics was

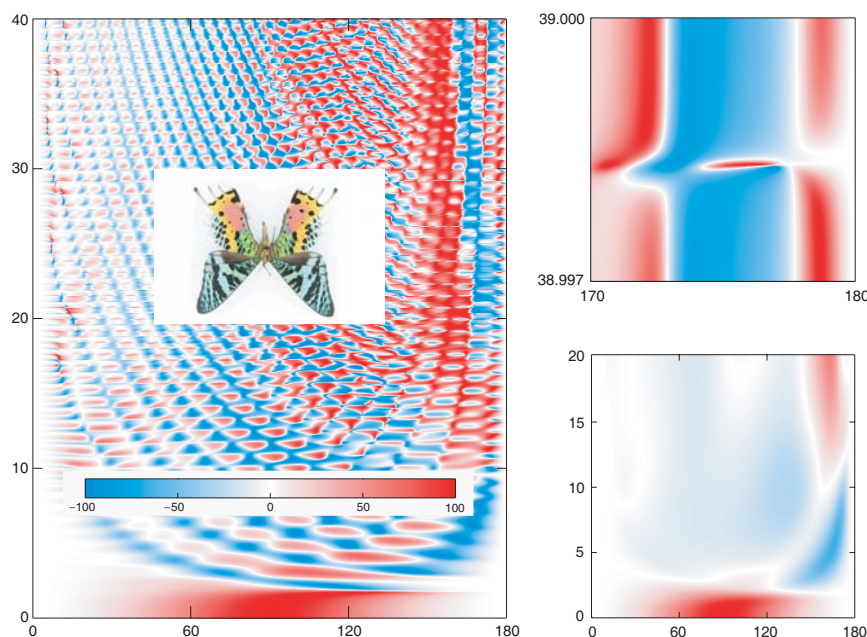


FIG. 5. Diagrams of the degree of linear polarization of scattered light in the case of unpolarized incident light for spherical particles with an index of refraction of (a), (b) 1.4, and (c) 1.44. The horizontal axes show the angle between the incidence and scattering directions (in degrees). The vertical axes in (a) and (b) show the dimensionless particle size parameter defined as the particle circumference divided by the wavelength of light; (c) computed for polydisperse spheres with a moderately wide size distribution, so the vertical axis shows the effective size parameter of the size distribution. The lower inset in (a) shows the color

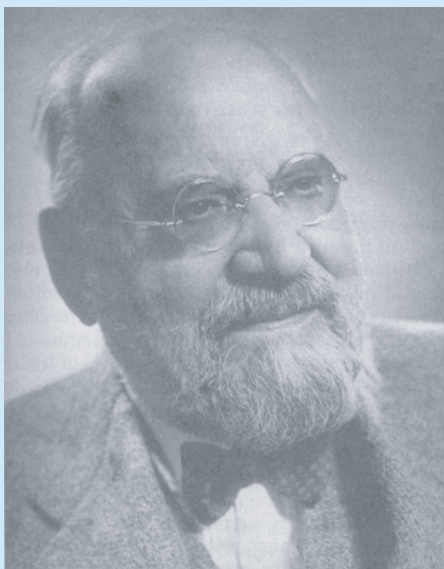
coding for the degree of linear polarization in percent. The upper inset in (a) is a photograph of the Madagascan sunset moth and illustrates why the specific distribution of red and blue spots in this polarization diagram is called the “butterfly structure.”

yet another striking demonstration of the practical power of the Mie theory.

CONCLUSIONS. The 1908 Mie paper was not the first one on the subject of wave scattering by a spherical particle. For example, Alfred Clebsch obtained the general solution of the elastic wave equation in terms of vector wave functions. The work of Clebsch was

consummated by Ludvig Lorenz, who also was the first to introduce what is now known as the Debye potentials. The latter were reinvented by Peter Debye, who also derived the famous formula for the radiation force exerted by the incident electromagnetic wave on a sphere. With the benefit of meticulously documented historical evidence, due credit should be given to these and several other prominent scientists who worked on

GUSTAV MIE (1868–1957)



Gustav Adolf Feodor Wilhelm Ludwig Mie was born on 29 September 1868 in Rostock, Germany, to the family of a merchant. He descended from Protestant pastor families on both his father's and mother's side. Growing up in a traditionally religious family environment undoubtedly affected his entire intellectual life. From 1886 Mie studied mathematics and physics at the University of Rostock. He also attended lectures in chemistry, zoology, geology, mineralogy, astronomy, logic, and metaphysics. In 1889 he continued his studies at the University of Heidelberg and received a doctoral degree in mathematics in 1891 with a dissertation "Zum Fundamentalsatz über die Existenz von Integralen partieller Differentialgleichungen" (On the fundamental existence theorem of integrals of partial differential equations). In 1897 Mie received *Habilitation* from the Karlsruhe Institute of Technology in theoretical physics.

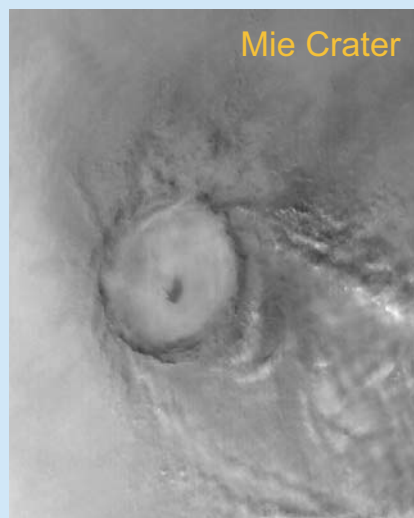
In 1901 he married Berta Hess (1875–1954) and a year later became an extraordinary professor of theoretical physics at the University of Greifswald, where he worked for 15 happy and scientifically productive years. At Greifswald he wrote the paper on electromagnetic scattering by homogeneous spherical particles that made him famous as well as his first book, *Molecules, Atoms, and Aether* (Mie 1904), which ran into four editions between 1904 and 1919. Interestingly, Mie's 1908 paper appears to be his single significant involvement with the subject of electromagnetic scattering by particles.

In 1917 Mie received and accepted an offer from the University of Halle, where he became a full professor of experimental physics and stayed until 1924. In the spring of 1924 he was invited to join the faculty of the University of Freiburg, where he worked as a professor until his formal retirement in 1935. Notably, Mie was a member of the Freiburger Kreise (Freiburg Circle), a group of economists, Catholics, and Protestants who met since 1938 at the University of Freiburg to develop concepts for a democratic post-Nazi Germany. Gustav Mie died on 13 February 1957 at the venerable age of 88.

Despite the eventually phenomenal success of his paper on electromagnetic scattering by spheres, Mie himself underestimated its importance, while taking special pride in his handbook on electromagnetics first published in 1910 and then revised and reprinted in 1941 and 1948 (Mie 1948a). Arnold Sommerfeld obviously agreed with Mie, characterizing this textbook as "excellent" (Sommerfeld 1952). More fundamentally, however, Mie was the first twentieth-century physicist to attempt a unified and complete theory of matter. Apparently it was this overarching quest that dwarfed in Mie's mind his previous particular work on electromagnetic scattering. Mie pursued his fundamental program in three "famous" (according to Sommerfeld) papers published in 1912 and 1913 (Mie 1912a,b, 1913). The importance of this contribution is well described by Mehra (1974) and Whittaker (1987). Subsequently, the great German mathematician David Hilbert used Mie's ideas to give the very first derivation of the equations of gravity (Hilbert 1915). It is, in fact, this derivation that is now in use in the majority of modern textbooks and monographs on general relativity and gravitation. In several essays written in the 1930s and 1940s Mie advocated a synthesis of Christian philosophy and natural sciences (e.g., Mie 1932).

In recognition of Mie's contributions to science, a 104-km-diameter crater on Mars as well as one of the buildings of the University of Freiburg were named after him.

More on Mie's life and personality can be found in his autobiography (Mie 1948b) as well as in the papers by Spehl (1990) and Lilienfeld (1991).



the wave scattering problem before or nearly simultaneously with Mie (Logan 1965; Kerker 1969). In particular, Lorenz's early contributions, which appear to have been unknown to Mie, have been acknowledged in the scientific literature in the last two decades by referring to the "Lorenz–Mie theory."

There is no doubt, however, that Mie's paper has had the most profound impact on the development of science, which is definitively documented by its fifth place on the all-time list of the most frequently cited papers in physics (Marx 2007). This happened because it provides

- a complete, comprehensive, and exact analytical solution of a classical scattering problem, including the case of an absorbing host medium, based on the fundamental principles of Maxwell's electromagnetics;
- a detailed and complete description of the numerical implementation of the analytical solution and a discussion of its significance in terms of partial electric and magnetic waves; and
- a convincing demonstration of the practical effectiveness of the numerical implementation via the presentation of actual tabular and graphical results and an application to the analysis of specific experimental data.

To quote Logan (1965), "Mie's paper is complete in itself, and it served as the basis for much of the work which has been done in this field since its publication."

The modern range of applications of the Mie theory is beyond comprehension. It includes such seemingly disparate disciplines as meteorological optics, biosensing with plasmonic nanoparticles (Shalaev and Kawata 2007), optical cloaking (Zhai et al. 2008), and the use of Mie MDRs to foster enhanced stimulated emission and thresholdless lasing (Fratilocchi et al. 2008). There is no doubt, however, that the full practical potential of the Mie theory is still to be revealed (see the sidebar for additional information about Gustav Mie).

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